

Static charge-imbalance effects in intrinsic Josephson systems

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Nonequilibrium effects created by stationary current injection in layered d-wave superconductors forming a stack of intrinsic Josephson junctions are studied. Starting from a nonequilibrium Green function theory we derive microscopic expressions for the charge-imbalance (difference between electron- and hole-like quasi-particles) on the superconducting layers and investigate its influence on the quasi-particle current between the layers. This nonequilibrium effect leads to shifts in the current-voltage curves of the stack. The theory is applied to the interpretation of recent current injection experiments in double-mesa structures.

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I. INTRODUCTION

In the strongly anisotropic cuprate superconductors like $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (BSCCO) the superconducting CuO_2 layers together with the intermediate insulating material form a stack of Josephson junctions. In the presence of a bias current perpendicular to the layers the intrinsic Josephson effect manifests itself in a multi-branch structure of the IV-curves,^{1,2,3,4,5} where on each branch a different number of junctions is in the resistive state.

In the resistive state high frequency Josephson oscillations at a finite dc-voltage are accompanied by a dc-current which is carried mainly by unpaired electrons (quasi-particles), while in the superconducting state it is carried mainly by Cooper pairs. Therefore, on a superconducting layer between a resistive and a superconducting junction the bias current has to change its character from quasi-particle current to supercurrent. This creates a non-equilibrium state on this layer with a finite quasi-particle charge and a change of the condensate charge. The quasi-particle charge is characterised by a distribution function with different numbers of electron-like and holelike quasi-particles, while the change of the condensate charge, the number of paired electrons, can be described by a shift of the chemical potential of the condensate.

In recent experiments such non-equilibrium effects have been observed in layered d-wave superconductors.⁶ In a first type of experiments Shapiro steps produced by high-frequency irradiation have been measured in mesa structures of BSCCO with gold contacts. Here a shift of the step voltage from its canonical value $\hbar f/(2e)$ was observed, which can be traced back to a change of the contact resistance due to quasi-particle charge on the first superconducting layer. In another type of experiments current-voltage curves have been investigated for two mesas structured close to each other on the same base crystal (see Fig. 7 below). Here an influence of the current through one mesa on the voltage drop through the other mesa has been measured which is caused by charge imbalance on the first common superconducting layer of the base crystal. In a recent paper⁷ we have ex-

plained these effects by using a semi-phenomenological approach based on a microscopic non-equilibrium Green function theory^{8,9} for layered superconductors.

In this paper we will present the full microscopic theory for stationary charge-imbalance effects in intrinsic Josephson systems. In order to be specific, we apply the theory first to an experiment, which in this form has not yet been done, but is conceptual simpler than the double-mesa injection experiment mentioned above. We consider a mesa as shown in Fig. 1 with two normal electrodes on top. Through one electrode a stationary bias current is applied creating a charge imbalance on the first superconducting layer. At the other electrode a voltage is measured as function of the bias current through the first electrode with zero current through the second electrode. This experiment is similar to the classical experiment by Clarke,¹⁰ where a strong current injected into a bulk superconductor creates quasi-particles. By two other electrodes, one normal junction and one Josephson junction, a voltage is detected at zero current which measures the difference between the chemical potential of quasi-particles and condensate. In our case the junction between the normal electrode and the first superconducting layer is the normal junction, the coupling between the first superconducting layer and the next superconducting layers is the Josephson junction. This experiment allows to determine the charge imbalance and (with further theoretical input) its relaxation rate. After this investigation we return to the discussion of the double-mesa injection experiment.

Nonequilibrium effects in *bulk-superconductors* have been studied a long time ago. For an overview see the text-book by Tinkham¹¹ and the review-articles edited by Langenberg and Larkin.¹² The basic concepts of charge-imbalance have been developed by Tinkham and Clarke,^{10,13} Pethick and Smith.¹⁴ The theory has been worked out using a non-equilibrium Green function approach by Schmid and Schön,^{15,16} and Larkin and Ovchinnikov.^{17,18} Microscopic nonequilibrium theory of layered superconductors was considered first by Artemenko¹⁹ and later by Graf et al.²⁰

Nonequilibrium effects in *intrinsic* Josephson system have been investigated theoretically already

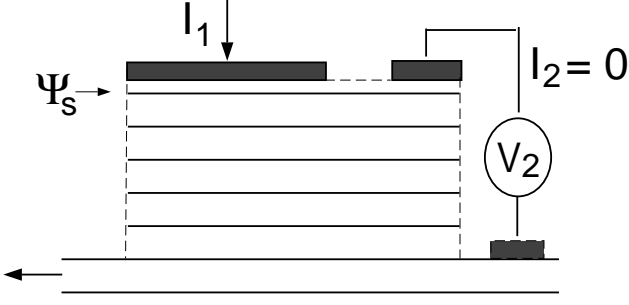


FIG. 1: Mesa-structure used for 4-point measurements consisting of a stack of superconducting layers with two normal electrodes on top.

in various contexts using different methods and approximations.^{21,22,23,24,25,26,27,28,29} In Refs. 21,28 the influence of charge-coupling on Josephson plasma oscillations has been investigated, in the approach used in Refs.^{23,24,26} a systematic perturbation theory in the scalar potential on the layers has been performed. In these theories charge imbalance is considered only indirectly as far as it is induced by fluctuations of the scalar potential. The present theory which uses non-equilibrium Green functions is more general. Here charge imbalance is taken into account as an independent degree of freedom, and therefore the results are different from those of earlier treatments.

Here we apply this theory to stationary processes in layered d-wave superconductors. Due to the weak coupling between the superconducting layers the total charge created by the bias-current is non-zero. Therefore we have to treat charge fluctuations of the condensate and quasi-particles as independent quantities. The d-wave character of the order-parameter allows quasi-particle tunnelling also at low voltages and temperatures. Furthermore it allows the relaxation of charge-imbalance by elastic scattering processes (impurity scattering within a layer). On the other hand the weak coupling between the layers leads to a partial decoupling of the kinetic equations for the Green functions in different layers, making this system a particularly simple example for the application of nonequilibrium theory in superconductors.

In the following section we start with general considerations concerning the Josephson effect in layered superconductors and the definition of charge-imbalance. We then formulate the basic kinetic equations describing stationary nonequilibrium effects in intrinsic Josephson systems and calculate the charge imbalance microscopically. We show how these effects can be measured by discussing the above mentioned 4-point experiment and the coupled two-mesa system. Finally we discuss the influence of charge-imbalance on the IV-curves of a stack of junctions.

II. GENERALISED JOSEPHSON EQUATIONS

We consider a system of superconducting layers with indices n and order parameter $\Delta_n(t) = |\Delta| \exp(i\chi_n(t))$ with time-dependent phase $\chi_n(t)$. We define the gauge invariant phase difference as:

$$\gamma_{n,n+1}(t) = \chi_n(t) - \chi_{n+1}(t) - \frac{2e}{\hbar} \int_n^{n+1} dz A_z(z, t), \quad (1)$$

where $A_z(z, t)$ is the vector potential in the barrier. Here e denotes the elementary charge. The charge of the electron is $-e$.

For the time derivative of $\gamma_{n,n+1}$ we obtain the generalized Josephson relation:

$$\frac{d\gamma_{n,n+1}}{dt} = \frac{2e}{\hbar} (V_{n,n+1} + \Phi_{n+1} - \Phi_n). \quad (2)$$

Here

$$V_{n,n+1} = \int_n^{n+1} dz E_z(z, t) \quad (3)$$

is the voltage and $\Phi_n(t)$ is the so-called gauge invariant scalar potential defined by

$$\Phi_n(t) = \phi_n(t) - \frac{\hbar}{2e} \dot{\chi}_n(t), \quad (4)$$

where $\phi_n(t)$ is the electrical scalar potential.

The quantity $\hbar \dot{\gamma}_{n,n+1} = 2e(V_{n,n+1} + \Phi_{n+1} - \Phi_n)$ is the total energy required to transfer a Cooper pair from layer n to $n+1$, $e\Phi_n$ can be considered as the shift of the chemical potential of the superconducting condensate with respect to an average chemical potential μ , i.e. the number of particles in the condensate is controlled by $\mu + e\Phi_n$. For equilibrium superconductors $\hbar \dot{\chi}_n = 2e\phi_n$, $\Phi_n = 0$, and one has the usual Josephson relation $\hbar \dot{\gamma}_{n,n+1} = 2eV_{n,n+1}$.

The generalised Josephson relation (2) for a stack of Josephson junctions can be written in another way showing explicitly the electric field distribution between the layers. We split the total charge fluctuation on a layer $\delta\rho_n = \delta\rho_n^c + \delta\rho_n^q$ into a contribution $\delta\rho_n^c$ from particles in the condensate and a contribution $\delta\rho_n^q$ from unpaired electrons (quasi-particles) describing charge-imbalance. The charge fluctuation of the condensate can be expressed directly by the change of the chemical potential on layer n :

$$\delta\rho_n^c = -2e^2 N(0) \Phi_n, \quad (5)$$

where $N(0)$ is the (two-dimensional) density of states for one spin direction at the Fermi energy. It is convenient to express also the fluctuation of quasi-particle charge by some quasi-particle potential Ψ_n , defining

$$\delta\rho_n^q = 2e^2 N(0) \Psi_n. \quad (6)$$

Then we obtain for the total charge density fluctuation:

$$\delta\rho_n = -2e^2 N(0) (\Phi_n - \Psi_n). \quad (7)$$

While in bulk superconductors charge neutrality leads to $\Phi = \Psi$, this is not the case in weakly coupled layered superconductors.

With help of (7) and the Maxwell equation (d is the distance between the layers)

$$\delta\rho_n = \frac{\epsilon\epsilon_0}{d}(V_{n,n+1} - V_{n-1,n}) \quad (8)$$

the generalized Josephson relation now reads:

$$\frac{\hbar}{2e}\dot{\gamma}_{n,n+1} = (1 + 2a)V_{n,n+1} - a(V_{n-1,n} + V_{n+1,n+2}) + \Psi_{n+1} - \Psi_n, \quad (9)$$

with $a = \epsilon\epsilon_0/(2e^2N(0)d)$. It shows that the Josephson oscillation frequency is determined not only by the voltage in the same junction but also by the voltages in neighboring junctions. The coefficient a has already been introduced by Koyama and Tachiki,²¹ however in their theory charge imbalance effects described by the quasi-particle potential Ψ_n have not been considered.

For a barrier in the superconducting state the time average $\langle\dot{\gamma}_{n,n+1}\rangle$ vanishes, while for a barrier in the resistive state $\hbar\langle\dot{\gamma}_{n,n+1}\rangle/2$ is the electrochemical potential difference. A similar equation also holds for the contact with the normal electrode. Denoting the normal electrode, which is assumed to be in equilibrium by $n = 0$, we have

$$\frac{\hbar}{2e}\dot{\gamma}_{0,1} = V_{0,1} + \Phi_1 = (1 + a)V_{0,1} + aV_{1,2} + \Psi_1. \quad (10)$$

In the case of a stack in contact with a normal electrode on top, where all internal barriers are in the superconducting state we find for the first superconducting layer $\Phi_1 \simeq aV + \Psi_1$. This result follows from Eqs. (2, 9, 10) in the limit $a \ll 1$. Note that the total voltage of the stack is $V = \sum_n \dot{\gamma}_{n,n+1}\hbar/(2e)$.

In order to describe transport between the layers we need also an expression for the current density. In our previous papers^{6,7} we have used

$$j_{n,n+1} = j_c \sin \gamma_{n,n+1} + \frac{\sigma_{n,n+1}}{d} \left(\frac{\hbar}{2e} \dot{\gamma}_{n,n+1} + \Psi_n - \Psi_{n+1} \right), \quad (11)$$

which is approximately valid in the stationary case (no displacement current). The first term is the current density of Cooper pairs. The rest is the quasi-particle current density, which is driven not only by the electrochemical potential difference, but contains a diffusion term proportional to the quasi-particle density difference. In the following the current expression will be derived from the microscopic theory.

Finally we need some theory describing the creation and relaxation of quasi-particle charge in order to calculate the charge-imbalance potential Ψ_n . In our previous papers we have used a relaxation time approximation. In the stationary case we obtained

$$\Psi_n = \tau_q(j_{n-1,n}^q - j_{n,n+1}^q)/[2e^2N(0)] \quad (12)$$

describing the balance between charge-imbalance creation by the in- and outgoing quasi-particle currents and relaxation into thermal equilibrium inside the layer. It will be the main task of this paper to derive such a relation from a microscopic theory.

III. KINETIC EQUATIONS

For the microscopic description we start from equations of motion for the spectral (retarded and advanced) and Keldysh Green functions in Nambu space (for details see the appendix A). Nonstationary corrections to the spectral functions are small, and these functions can be taken in equilibrium. The Keldysh function contains the necessary information about the nonequilibrium distribution functions. In our model we consider d-wave superconductivity on each layer within the BCS-approximation, elastic impurity scattering inside the layer and tunnelling between neighboring layers in lowest order. Then it is possible to formulate equations of motion for the Green function $\hat{G}_n^K(\vec{k}, t_1, t_2)$ for each layer n with a tunnelling self-energy containing the coupling to the neighboring layers.

Furthermore we introduce quasiclassical Green functions, which are obtained by integrating the Green function over the kinetic energy $\xi_k = \epsilon_k - \mu$. Performing in addition a Fourier transformation with respect to the time difference $\tau = t_1 - t_2$ at fixed central time $t = (t_1 + t_2)/2$ we obtain (see the appendix B) the following kinetic equation for the quasiclassical Keldysh function $\hat{g}_n^K(\vec{k}, t, \epsilon)$ in Nambu space:

$$\begin{aligned} \left(\frac{i}{2} \frac{\partial}{\partial t} + \epsilon \right) \tau_3 \hat{g}^K + \left(\frac{i}{2} \frac{\partial}{\partial t} - \epsilon \right) \hat{g}^K \tau_3 \\ - \{ \hat{h}(\vec{k}, t) \hat{g}^K - \hat{g}^K \hat{h}(\vec{k}, t) \} \\ = \{ \hat{\sigma}^R \hat{g}^K + \hat{\sigma}^K \hat{g}^A - \hat{g}^R \hat{\sigma}^K - \hat{g}^K \hat{\sigma}^A \} \end{aligned} \quad (13)$$

depending on the central time t , the direction \hat{k} of the momentum, and the frequency ϵ . Here

$$\begin{aligned} \hat{h}(\vec{k}, t) &= -e\Phi_n(t) - \hat{\Delta}(\vec{k}), \\ \hat{\Delta}(\vec{k}) &= \begin{pmatrix} 0 & \Delta(\vec{k}) \\ -\Delta(\vec{k}) & 0 \end{pmatrix}, \end{aligned} \quad (14)$$

where $\Phi_n(t)$ is the gauge invariant scalar potential Eq.(4). $\Delta(\vec{k})$ is the superconducting order parameter with d-wave symmetry. We have applied a gauge transformation such that on each layer the superconducting order parameter has the same phase (in our case it is real). The quantities $\hat{\sigma}$ are self-energies due to impurity scattering and tunnelling, which will be specified later. The curly brackets denote a convolution in time and frequency space. In the stationary case, which we consider in the following, we will neglect the time dependence. Then the convolutions are simple products, the depen-

dence on $\Phi_n(t)$ drops out, and we obtain the basic equation

$$\begin{aligned} \epsilon(\tau_3 \hat{g}^K - \hat{g}^K \tau_3) + \hat{\Delta} \hat{g}^K - \hat{g}^K \hat{\Delta} = \\ = [\hat{\sigma}^R \hat{g}^K + \hat{\sigma}^K \hat{g}^A - \hat{g}^R \hat{\sigma}^K - \hat{g}^K \hat{\sigma}^A]. \end{aligned} \quad (15)$$

This is actually an equation for the nonequilibrium distribution functions contained in the Keldysh Green function \hat{g}^K and the Keldysh selfenergy $\hat{\sigma}_K$.

In the following we will use the ansatz by Larkin and Ovchinnikov:^{17,18}

$$\hat{g}^K = \{\hat{g}^R \hat{d} - \hat{d} \hat{g}^A\} \quad (16)$$

with

$$\hat{d} = \beta + \alpha \hat{\tau}_3$$

containing two distribution functions β and α . In the stationary nonequilibrium case we can neglect the convolution and replace it by products:

$$\hat{g}^K = (\hat{g}^R - \hat{g}^A)\beta + (\hat{g}^R \tau_3 - \tau_3 \hat{g}^A)\alpha, \quad (17)$$

where for the retarded and advanced functions $\hat{g}^{R,A}(\epsilon)$ the equilibrium functions can be used. In equilibrium

$$\beta(\epsilon) = \tanh(\epsilon/(2T)), \quad \alpha(\epsilon) = 0. \quad (18)$$

Returning now to the equation of motion Eq.(15) and using the traditional notation (see appendix A) for matrices in Nambu space, $g_{11} =: g, g_{12} =: f, g_{21} =: -f^+, g_{22} =: \bar{g}$, the stationary part of the equations of motion for the normal and anomalous Keldysh Green functions read explicitly

$$-\Delta(f^{+K} - f^K) = I_{11}, \quad (19)$$

$$+\Delta(f^{+K} - f^K) = I_{22}, \quad (20)$$

$$2\epsilon f^K + \Delta(\bar{g}^K - g^K) = I_{12}, \quad (21)$$

$$2\epsilon f^{+K} + \Delta(\bar{g}^K - g^K) = I_{21}, \quad (22)$$

with the abbreviation for the tunnelling and scattering integrals:

$$I_{\alpha\beta} = [\hat{\sigma}^R \hat{g}^K + \hat{\sigma}^K \hat{g}^A - \hat{g}^R \hat{\sigma}^K - \hat{g}^K \hat{\sigma}^A]_{\alpha\beta}.$$

As we shall see later $\langle f \rangle = \langle \sigma_{12} \rangle = \langle \sigma_{21} \rangle = 0$ for d-wave symmetry, and we obtain:

$$\begin{aligned} I_{11} &= \sigma_{11}^R g^K - g^R \sigma_{11}^K + \sigma_{11}^K g^A - g^K \sigma_{11}^A \\ I_{22} &= \sigma_{22}^R \bar{g}^K - \bar{g}^R \sigma_{22}^K + \sigma_{22}^K \bar{g}^A - \bar{g}^K \sigma_{22}^A \\ I_{12} &= \sigma_{11}^R f^K - f^R \sigma_{22}^K + \sigma_{11}^K f^A - f^K \sigma_{22}^A \\ I_{21} &= -\sigma_{22}^R f^{+K} + f^{+R} \sigma_{11}^K - \sigma_{22}^K f^{+A} + f^{+K} \sigma_{11}^A \end{aligned} \quad (23)$$

The scattering terms on the r.h.s have different meaning: those which are proportional to the function itself can be considered as self-energy. Therefore we add these terms to the l.h.s. of Eqs.(21) and (22), writing:

$$2\bar{\epsilon} f^K + \Delta(\bar{g}^K - g^K) = J_{12} \quad (24)$$

with

$$\bar{\epsilon} = \epsilon - \frac{1}{2}(\sigma_{11}^R + \sigma_{11}^A) = \epsilon - \text{Re}\sigma_{11}^R \quad (25)$$

where we used the equilibrium values for $\sigma_{11}^{R,A} = -\sigma_{22}^{R,A}$, and $\sigma_{11}^A = (\sigma_{11}^R)^*$. Then the correction to ϵ is just the real part of the scattering self-energy evaluated below. The scattering term on the r.h.s. is then

$$\begin{aligned} J_{12} &= -f^R \sigma_{22}^K + \sigma_{11}^K f^A, \\ J_{21} &= f^{+R} \sigma_{11}^K - \sigma_{22}^K f^{+A} \end{aligned} \quad (26)$$

Now we substitute the anomalous Keldysh functions f^K and f^{+K} in Eqs.(19, 20) and obtain

$$I_{11} - I_{22} - \frac{\Delta}{\bar{\epsilon}}(J_{12} - J_{21}) = 0 \quad (27)$$

$$I_{11} + I_{22} = 0 \quad (28)$$

These equations will be used in the following to determine the distribution functions $\alpha(\epsilon)$ and $\beta(\epsilon)$ describing the non-equilibrium state. In particular, Eq.(27) describes the balance between the relaxation of quasi-particle charge due to impurity scattering within a layer and its creation by the tunnelling current (see Eq.(43) for the result in a special case).

IV. CALCULATION OF THE CHARGE IMBALANCE

We consider potential scattering from randomly distributed impurity centers and tunnelling to the neighboring layers in lowest order. In order to calculate the self-energies and scattering integrals we need expressions for the retarded and advanced Green functions. Here we use the results from equilibrium (this can be justified by studying the corresponding equations of motion in the low frequency limit).

A. Retarded and advanced Green functions

The retarded (advanced) quasi-classical Green functions in equilibrium have the form:

$$\begin{aligned} g^R(\hat{k}, \epsilon) &= \frac{\epsilon + i\gamma_\epsilon}{\sqrt{(\epsilon + i\gamma_\epsilon)^2 - \Delta^2(\hat{k})}}, \quad g^A = -(g^R)^*, \\ f^R(\hat{k}, \epsilon) &= \frac{\Delta(\hat{k})}{\sqrt{(\epsilon + i\gamma_\epsilon)^2 - \Delta^2(\hat{k})}}, \quad f^A = -(f^R)^*, \end{aligned} \quad (29)$$

with $-i\gamma_\epsilon = \sigma_{11}^R(\epsilon)$. Here we use the sign-convention $\text{Im}\sqrt{} > 0$. Furthermore we have for the equilibrium

functions $f^{+R,A} = f^{R,A}$, $\bar{g}^{R,A} = -g^{R,A}$. In the following we will also need the combinations:

$$\begin{aligned} u(\hat{k}, \epsilon) &= \frac{1}{2}(g^R(\hat{k}, \epsilon) - g^A(\hat{k}, \epsilon)), \\ v(\hat{k}, \epsilon) &= \frac{1}{2}(f^R(\hat{k}, \epsilon) - f^A(\hat{k}, \epsilon)), \\ w(\hat{k}, \epsilon) &= \frac{i}{2}(f^R(\hat{k}, \epsilon) + f^A(\hat{k}, \epsilon)). \end{aligned} \quad (30)$$

The (even) spectral function $u(\hat{k}, \epsilon)$ is the tunnelling density of states. With this notation we obtain for the Keldysh functions Eq.(17)

$$\begin{aligned} g^K(\hat{k}, \epsilon) &= 2u(\hat{k}, \epsilon)(\alpha(\hat{k}, \epsilon) + \beta(\hat{k}, \epsilon)), \\ \bar{g}^K(\hat{k}, \epsilon) &= 2u(\hat{k}, \epsilon)(-\beta(\hat{k}, \epsilon) + \alpha(\hat{k}, \epsilon)). \end{aligned} \quad (31)$$

B. Potential scattering

The self-energies in Born approximation are given by:

$$\hat{\sigma}_p^{R,A,K}(\epsilon) = c \sum_{k'} |V_0|^2 \hat{G}^{R,A,K}(\vec{k}, \epsilon) = -i\nu_p \langle \hat{g}^{R,A,K}(\epsilon) \rangle \quad (32)$$

with $\nu_p = c\pi N(0)|V_0|^2$. Here V_0 is the scattering potential and c the impurity concentration. For comparison in the numerical results we also use the t-matrix approximation in the strong scattering limit. This is obtained by replacing ν_p by $\nu_p/|g^R(\epsilon)|^2$.

For the calculation of the Keldysh component of the self-energy we use the ansatz by Larkin and Ovchinnikov Eq.(17) and obtain

$$\sigma_p^K(\epsilon) = -2i\nu_p \langle u(\epsilon)\beta(\epsilon) \rangle \tau_3 - 2i\nu_p \langle v(\epsilon)\alpha(\epsilon) \rangle \tau_0. \quad (33)$$

Using these results we obtain for the potential part of the scattering terms:

$$\begin{aligned} I_{11}^p &= -I_{22}^p = 4i\nu_p [u(\epsilon) \langle u(\epsilon)\alpha(\epsilon) \rangle - \langle u(\epsilon) \rangle u(\epsilon)\alpha(\epsilon)], \\ J_{12}^p - J_{21}^p &= 8i\nu_p v(\epsilon) \langle u(\epsilon)\alpha(\epsilon, t) \rangle. \end{aligned} \quad (34)$$

C. Tunneling

We describe the coupling between neighboring layers by the usual tunnelling Hamiltonian

$$H = \sum_{n\sigma k k'} (t_{k'k} e^{-\frac{2e}{\hbar} \int_n^{n+1} dz A_z(z, t)} c_{n+1, k'\sigma}^\dagger c_{n, k\sigma} + h.c.), \quad (35)$$

which after the gauge transformation becomes

$$H = \sum_{n\sigma k k'} (t_{k'k} e^{i\gamma_{n,n'}(t)} c_{n+1, k'\sigma}^\dagger c_{n, k\sigma} + h.c.). \quad (36)$$

Then we obtain for the contribution to the tunnelling self-energy from the coupling between layers n and $n' = n \pm 1$:

$$\begin{aligned} \hat{\sigma}_t(\epsilon, t)_{n,n'} &= \int d(t_1 - t_2) e^{i\epsilon(t_1 - t_2)} \sum_{k'} |t_{k'k}|^2 \\ &\quad e^{-i\tau_3 \gamma_{n,n'}(t_1)/2} \hat{G}(\vec{k}', t_1, t_2) e^{+i\tau_3 \gamma_{n,n'}(t_2)/2}. \end{aligned} \quad (37)$$

If we neglect additional phase fluctuation, the phase difference is $\gamma_{n,n'}(t) = \gamma_{n,n'}^0 + \Omega_{n,n'} t$, where $\gamma_{n,n'}^0$ is the static phase difference, which is determined by the supercurrent, and $\Omega_{n,n'}$ is proportional to the electrochemical potential difference between the two layers.

For the tunnelling matrix element we have to assume a partial conservation of momentum in order to obtain Josephson coupling between different superconducting layers with d-wave order parameter. This, however, is not relevant for the tunnelling of quasiparticles, which will be discussed here primarily. In the following we will model the tunnelling matrixelement near the Fermi surface by $\pi N(0)|t_{k'k}|^2 = \nu_t(\hat{k}, \hat{k}')$ with $\langle \Delta(\hat{k}) \nu_t(\hat{k}, \hat{k}') \Delta(\hat{k}') \rangle \neq 0$.

The most simple case to study nonequilibrium effects is the tunnelling between a normal electrode and a superconducting layer, which will be discussed in the following. Results for quasi-particle tunnelling between two superconducting layers will be given in the appendix. In the case of tunnelling with a normal electrode only the normal Green functions contribute to the tunnelling self-energy, which is time-independent. Writing $v = \Omega_{n,n'}/2$ we obtain

$$\begin{aligned} \hat{\sigma}_t^R(\hat{k}, \epsilon)_{(n,n')} &= -\frac{i}{2} \left(\langle \nu_t g^R(\epsilon - v) \rangle' - \langle \nu_t g^R(\epsilon + v) \rangle' \right) \tau_0 \\ &\quad - \frac{i}{2} \left(\langle \nu_t g^R(\epsilon - v) \rangle' + \langle \nu_t g^R(\epsilon + v) \rangle' \right) \tau_3, \end{aligned} \quad (38)$$

$$\begin{aligned} \hat{\sigma}_t^K(\hat{k}, \epsilon)_{(n,n')} &= \\ &\quad -i \left(\langle \nu_t u'(\epsilon - v) \beta'(\epsilon - v) \rangle' - \langle \nu_t u'(\epsilon + v) \beta'(\epsilon + v) \rangle' \right) \tau_0 \\ &\quad -i \left(\langle \nu_t u'(\epsilon - v) \beta'(\epsilon - v) \rangle' + \langle \nu_t u'(\epsilon + v) \beta'(\epsilon + v) \rangle' \right) \tau_3. \end{aligned} \quad (39)$$

The prime denotes spectral functions and distribution functions on layer n' and $\langle \nu_t g^R \rangle' = \langle \nu_t(\hat{k}, \hat{k}') g^R(\hat{k}', \epsilon) \rangle_{k'}$ denotes an average over the direction of \hat{k}' .

These results will be used to calculate the tunnelling contributions to the scattering terms (27,28) in the kinetic equation. Denoting the superconducting layer by n , the normal layer by n' , replacing the spectral function on the normal layer by $u'(\epsilon) = 1$, and neglecting terms of order $\nu_t \alpha$ we find

$$\begin{aligned} I_{11}^{tn} - I_{22}^{tn} &= 4i \langle \nu_t (\beta'(\epsilon - v) - \beta'(\epsilon + v)) u(\epsilon) \rangle', \\ I_{11}^{tn} + I_{22}^{tn} &= 4i \langle \nu_t (\beta'(\epsilon - v) + \beta'(\epsilon + v) - \beta(\epsilon) u(\epsilon)) \rangle', \\ J_{12}^{tn} - J_{21}^{tn} &= 4i \langle \nu_t v(\epsilon) (\beta'(\epsilon - v) - \beta'(\epsilon + v)) \rangle'. \end{aligned} \quad (40)$$

In order to determine the charge-imbalance on the superconducting layer in the stack we also need the quasi-particle contribution from tunnelling into the neighboring

superconducting layer with the barrier in the superconducting state. As will be shown in the Appendix, this contribution vanishes if we neglect terms of order $\nu_t \alpha$.

D. Solution of the kinetic equation

Inserting now the different contributions to the scattering term from potential scattering Eq.(34) and tunnelling Eq.(40) into Eq.(27) we obtain the following equation determining the distribution function $\alpha(\hat{k}, \epsilon)$ on the superconducting layer:

$$\begin{aligned} & 2\nu_p [u(\epsilon) \langle u(\epsilon) \alpha(\epsilon) \rangle - \langle u(\epsilon) \rangle u(\epsilon) \alpha(\epsilon)] \\ & - 2\nu_p \frac{\Delta v(\epsilon)}{\tilde{\epsilon}} \langle u(\epsilon) \alpha(\epsilon) \rangle \\ & = - \left(u(\epsilon) - \frac{\Delta}{\tilde{\epsilon}} v(\epsilon) \right) \langle \nu_t(\hat{k}, \hat{k}') [\beta'(\epsilon - v) - \beta'(\epsilon + v)] \rangle'. \end{aligned} \quad (41)$$

The r.h.s. of this equation describes charge imbalance generation by tunnelling of quasi particles. The l.h.s. describes charge imbalance relaxation due to impurity scattering. Taking an angular average and defining

$$\begin{aligned} \tilde{\alpha}(\epsilon) &:= \langle u(\epsilon) \alpha(\epsilon) \rangle, \\ R(\epsilon) &:= u(\epsilon) - \frac{\Delta v(\epsilon)}{\tilde{\epsilon}}, \\ \nu(\epsilon) &:= 2\nu_p \left\langle \frac{\Delta v(\epsilon)}{\tilde{\epsilon}} \right\rangle, \end{aligned} \quad (42)$$

we obtain

$$\tilde{\alpha}(\epsilon) = \frac{\langle \nu_t R(\epsilon) \rangle}{\nu(\epsilon)} (\beta'(\epsilon - v) - \beta'(\epsilon + v)). \quad (43)$$

Finally from Eq.(28) and Eq.(40) we obtain for the distribution function on the superconducting layer

$$\beta(\epsilon) = \frac{1}{2} (\beta'(\epsilon - v) + \beta'(\epsilon + v)), \quad (44)$$

where $\beta'(\epsilon)$ is the distribution function on the normal layer, which is assumed to be in equilibrium. This simple relation is only true as long as we neglect inelastic scattering, which is necessary for a relaxation of quasi-particles into the condensate.

E. Charge-imbalance

Equation (43) is the main result of the paper. The function $\tilde{\alpha}(\epsilon)$ describes the charge of quasi-particles with energy ϵ on the superconducting layer. It is related to the quasi-particle potential Ψ introduced in the phenomenological theory in the following way: The quasiclassical expression for the charge density is

$$\delta\rho_n = -2eN(0) \left[e\Phi_n + \int_{-\infty}^{\infty} \frac{d\epsilon}{4} \langle g_n^K(\hat{k}, \epsilon) \rangle \right]. \quad (45)$$

Using the distribution function α , introduced in the previous section, we can write this expression as

$$\begin{aligned} \delta\rho_n &= -2eN(0) \left[e\Phi_n - \int_0^{\infty} d\epsilon \langle u(\hat{k}, \epsilon) \alpha(\hat{k}, \epsilon) \rangle \right] \\ &= -2e^2 N(0) [\Phi_n - \Psi_n], \end{aligned} \quad (46)$$

with the charge-imbalance potential determined by the formula

$$\Psi = (1/e) \int_0^{\infty} d\epsilon \langle u(\hat{k}, \epsilon) \alpha(\hat{k}, \epsilon) \rangle. \quad (47)$$

Relation of $\alpha(\hat{k}, \epsilon)$ to the "clean limit" charge-imbalance distribution function α_k in k-space is given in Appendix D. The relaxation rate $\nu(\epsilon)$ describes the relaxation into thermal equilibrium of the difference between electron and hole-like quasi-particles due to impurity scattering. It replaces the relaxation rate $1/\tau_q$ in Eq.(12). The r.h.s. of Eq.(43) contains the tunnelling of quasi-particle charge from the normal electrode into the superconducting layer due to the applied voltage. It is similar but not equal to the tunnelling current at the same energy ϵ . In the tunnelling current (see below) the function $R(\epsilon)$ is replaced by the density-of-states function $u(\epsilon)$. In the absence of impurity scattering one finds $R(\epsilon) = 1/u(\epsilon)$ in agreement with Ref.13.

V. CALCULATION OF THE TUNNELLING CURRENT

In order to measure the charge-imbalance induced by the quasi-particle injection we also need an expression for the current, in particular between the normal contact and the first superconducting layer. Quite generally the current between neighboring layers n and n' in lowest order in the tunnelling matrix element can be written as

$$\begin{aligned} J_{n,n'}(t) &= + \frac{2e}{\hbar} \sum_{kk'} \int dt_1 |t_{kk'}|^2 \\ & \left(\hat{G}_n^R(k, t, t_1) e^{-i\tau_3 \gamma_{n,n'}(t_1)/2} \hat{G}_{n'}^<(k, t_1, t) e^{+i\tau_3 \gamma_{n,n+1}(t)/2} \right. \\ & \left. \hat{G}_n^<(k, t, t_1) e^{-i\tau_3 \gamma_{n,n'}(t_1)/2} \hat{G}_{n'}^A(k, t_1, t) e^{+i\tau_3 \gamma_{n,n'}(t)/2} \right)_{11} \\ & + c.c. \end{aligned} \quad (48)$$

In the stationary case (constant applied voltage) we neglect the time dependence of the Green functions on the central time and use a Fourier transformation with respect to the time difference as defined above. Furthermore we restrict ourselves to the tunnelling between the normal layer and the first superconducting layer. Then only the normal Green functions contribute and we obtain (with $v = \Omega_{n,n'}/2$) the usual expression for the

quasi-particle current density

$$j_{n,n'}(v) = -\frac{e}{\hbar} N(0) \int_{-\infty}^{+\infty} d\epsilon \left(\langle \nu_t(\hat{k}, \hat{k}') g^R(\hat{k}, \epsilon) g^<(\hat{k}', \epsilon - v) \rangle + \langle \nu_t(\hat{k}, \hat{k}') g^<(\hat{k}, \epsilon) g^A(\hat{k}', \epsilon - v) \rangle \right) + c.c. \quad (49)$$

Here the prime denotes momenta and distribution functions on layer n' , and the double brackets denote an average over both \hat{k} and \hat{k}' .

Now we express the lesser functions $g^<$ by the Keldysh functions

$$\hat{g}^< = \frac{1}{2}(\hat{g}^K - \hat{g}^R - \hat{g}^A) \quad (50)$$

and express the latter by the non-equilibrium distribution functions β and α , then

$$j_{n,n'}(v) = -\frac{2e}{\hbar} N(0) \int_{-\infty}^{+\infty} d\epsilon \langle \nu(\hat{k}, \hat{k}') u(\hat{k}, \epsilon) u'(\hat{k}', \epsilon - v) [\beta'(\epsilon - v) - \beta(\epsilon) + \alpha'(\epsilon - v) - \alpha(\epsilon)] \rangle \\ =: j_{n,n'}^\beta(v) + j_{n,n'}^\alpha(v). \quad (51)$$

This is another important result. The current between neighboring layers n and n' is the sum of two parts containing the distribution functions $\beta(\epsilon)$ and $\alpha(\epsilon)$ respectively. The current $j^\beta(v)$ is the quasi-particle current driven and created by the electro-chemical potential difference between the two layers. The current $j^\alpha(v)$ describes the diffusion current driven by the charge imbalance. Both current contributions depend on the density of state $u(\epsilon)$ of the two layers.

For the further application to the tunnelling between a normal electrode and a superconducting layer it is convenient to denote by j the current flow *from* the normal electrode *to* the superconducting layer and by V the voltage drop in this direction (i.e. $v = -eV$). With $u'(\epsilon) = 1$, $\alpha' = 0$, $\beta'(\epsilon) = \beta_0(\epsilon) = \tanh(\epsilon/(k_B T))$ for the normal layer, and exploiting the fact that $u(\epsilon)$ and $\alpha(\epsilon)$ are even functions, while $\beta(\epsilon)$ is an odd function of ϵ we obtain for the current contributions:

$$j^\beta(V) = \frac{e}{\hbar} N(0) \nu_t \int_{-\infty}^{+\infty} d\epsilon \langle u(\hat{k}, \epsilon) [\beta_0(\epsilon + eV) - \beta_0(\epsilon - eV)] \rangle =: j_0(V), \quad (52)$$

$$j^\alpha(V) = -\frac{2e}{\hbar} N(0) \nu_t \int_{-\infty}^{+\infty} d\epsilon \langle u(\hat{k}, \epsilon) \alpha(\hat{k}, \epsilon) \rangle \\ =: -\sigma_0 \Psi(V). \quad (53)$$

Here we have replaced the tunnelling rate by some average $\nu_t = \langle \nu(\hat{k}, \hat{k}') \rangle$ and have defined the ohmic resistance $\sigma_0 = 4e^2 N(0) \nu_t / \hbar$. Then the current driven by the nonequilibrium distribution of quasi-particles can be

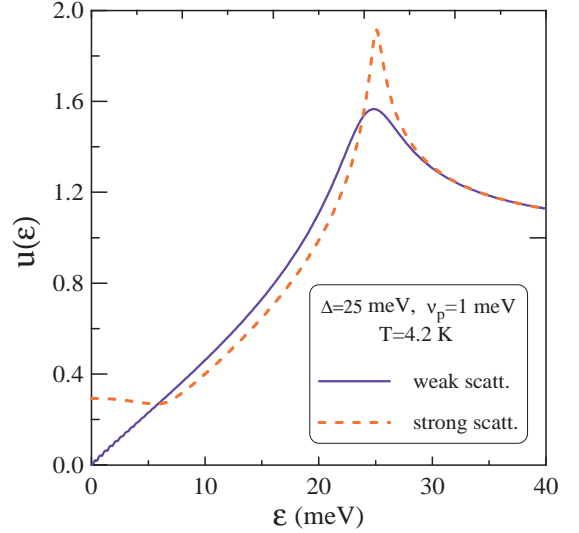


FIG. 2: Spectral function (tunnelling density of states) calculated for a d-wave superconductor with impurity scattering.

expressed by the quasi-particle potential

$$\Psi(V) = \frac{1}{e} \int_0^\infty d\epsilon \langle u(\hat{k}, \epsilon) \alpha(\hat{k}, \epsilon) \rangle \\ = \frac{1}{e} \int_0^\infty d\epsilon \frac{\nu_t}{\nu(\epsilon)} \langle R(\epsilon) \rangle [\beta_0(\epsilon + eV) - \beta_0(\epsilon - eV)]. \quad (54)$$

VI. RESULTS

For our model calculations we use a d-wave order parameter with an angular dependence of the usual form $\Delta(\hat{k}) = \Delta(T) \phi(\hat{k})$ with $\phi(\hat{k}) = \cos 2\theta_k$. The tunnelling matrix element will be parametrised as $\nu_t(\hat{k}, \hat{k}') = \nu_1 + \nu_2 \phi(\hat{k}) \phi(\hat{k}')$. This simplifies the averaging procedure: the parameter ν_1 enters the normal tunnelling probability while the parameter ν_2 determines the Josephson coupling.

A. Tunneling current and charge imbalance

In Fig. 2 we show well-known typical results for the spectral function (tunnelling density of states) $u(\epsilon)$ at low temperatures with a self-consistently determined self-energy $i\gamma(\epsilon)$ for the two limiting cases of Born scattering and in the unitary limit. Note that in the unitary limit the spectral function stays finite for $\epsilon \rightarrow 0$. This function will be needed as input for the following calculations of the tunnelling current and the charge-imbalance relaxation. Fig. 3 shows the corresponding normalized tunnelling current $j_0(V)/\sigma_0$ calculated from Eq.(52) between a normal electrode and a superconducting layer as function of the voltage-drop V between the normal electrode and the superconducting layer.

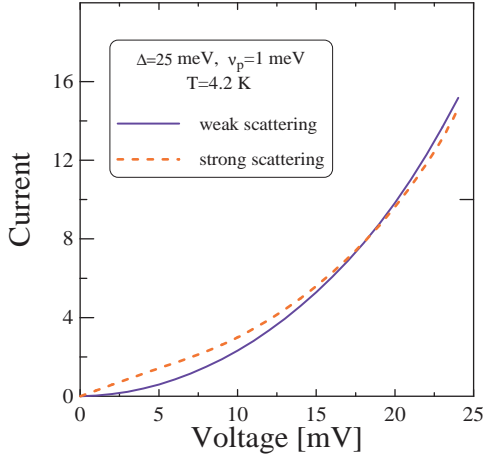


FIG. 3: Tunnelling current j_0/σ_0 [mV] as function of the applied voltage.

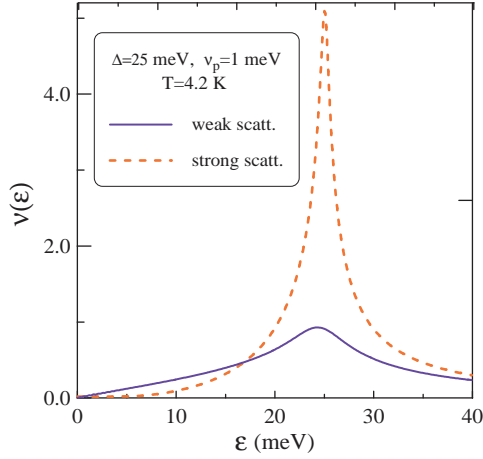


FIG. 4: Relaxation rate for the charge-imbalance due to impurity scattering as function of energy.

Now we turn to the calculation of charge imbalance. In Fig. 4 the frequency dependence of the relaxation function $\nu(\epsilon)$ is shown for low temperatures. The frequency dependence reflects the available phase-space for elastic scattering processes from electron-like into hole-like quasi-particles at the energy ϵ . With help of this function we calculate from Eq.(54) the charge imbalance potential Ψ generated on a superconducting layer in contact with a normal layer. Fig. 5a shows $\Psi(V)$ as function of the voltage V between the normal electrode and the superconducting layer.

As V cannot easily be measured directly, we express it by the corresponding current density. The resulting function $\psi(j)$ defined by $\psi(j) = \Psi(V)$ with $j = j_0(V)$ given by Eq.(52) is shown in Fig. 5b. It depends on the ratio ν_t/ν_p between the average tunnelling rate and the potential scattering rate. The nonlinear dependence on the current reflects to some extent the nonlinear current

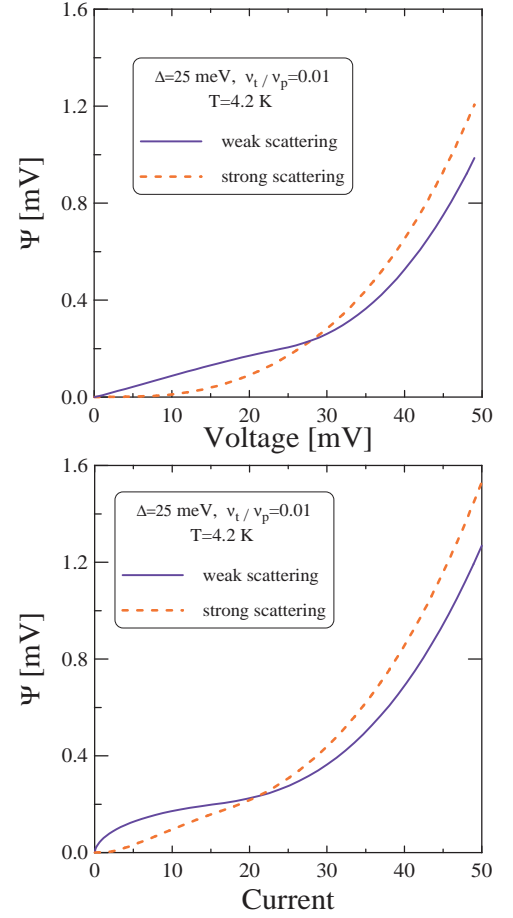


FIG. 5: Charge imbalance potential generated on a superconducting layer in contact with a normal electrode as function of the applied voltage (a) and the tunnelling current j_0/σ_0 (b)

voltage curve.

B. Experiment with two normal electrodes

Now we apply the theory to the basic experiment, where one superconducting layer is in contact with two normal electrodes. Through the first electrode with area F_1 a current I_1 is applied, which creates a charge imbalance Ψ_s on the superconducting layer. At a second electrode the voltage V_2 is measured with no current flowing. We assume that charge imbalance spreads evenly over the whole superconducting layer of size F . Then the charge imbalance potential created on the superconducting layer is given by $\Psi_s = (F_1/F)\psi(j_1)$, where $j_1 = I_1/F_1$ is the current density through the electrode (1), with $\psi(j)$ defined above and shown in Fig. 5b.

In order to determine the voltage V_2 measured at the second electrode we use the current equations (52, 53) and exploit the condition that no current is flowing, $j_2 = j_0(V_2) - \sigma_0\Psi_s = 0$, i.e. we have a compensation of the quasi-particle current driven by the voltage and

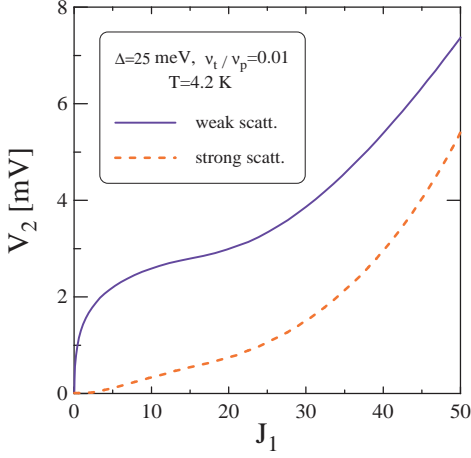


FIG. 6: Voltage measured at the second electrode as function of the injection current through the first electrode.

the quasi-particle diffusion current driven by the charge imbalance potential. Using for Ψ_s the value determined above we find

$$\int_0^\infty d\epsilon \langle u(\hat{k}, \epsilon) [\beta_0(\epsilon + eV_2) - \beta_0(\epsilon - eV_2)] \rangle = \frac{F_1}{F} \psi(j_1). \quad (55)$$

Thus we obtain V_2 as function of j_1 . Results are shown in Fig. 6 for the case of a very small test electrode, $F_2 \ll F$, $F_1 \simeq F$. Note that in this channel the electrochemical potential drop is only between the normal electrode and the first superconducting layer, all the other barriers in the stack have $\Omega_{n,n+1} = 0$. Furthermore, if the electrodes are in equilibrium, the total voltage equals the total electrochemical potential, thus V_2 is the total measured voltage.

C. Double-mesa experiment

In this experiment which is described in detail in Ref.6 two small mesas are structured close to each other on top of a common base mesa and contacted with separate gold electrodes. Through the first mesa a variable current I_1 is injected while at the second mesa the voltage V_2 is measured for fixed current I_2 . Normally the voltage V_2 is independent of the current I_1 as long as all junctions in the base mesa are in the superconducting state. In some cases, however, a small additional voltage $\Delta V_2(I_1)$ is observed. This happens, if the lowest junctions in the two mesas are in the resistive state (see Fig. 7b) and generate a charge-imbalance on the first common superconducting layer of the base mesa, which then depends on both currents.

If we want to apply the microscopic theory to this situation we have to calculate the charge-imbalance on a superconducting layer in contact with another superconducting layer with the barrier being in the resistive state. Furthermore we have to add to the tunnelling current the

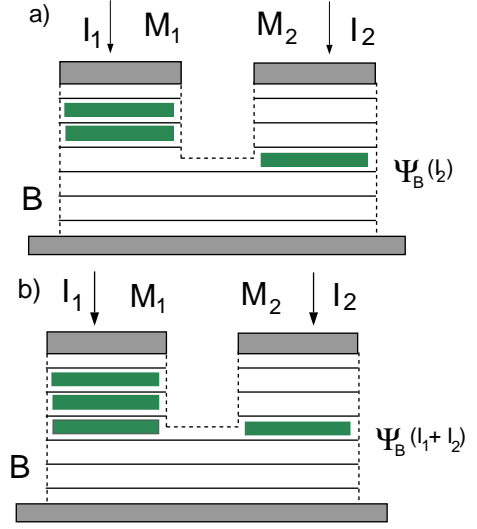


FIG. 7: Position of resistive barriers in the double-mesa experiment

average Josephson current. In systems with a large McCumber parameter this contribution is small. We will neglect it in the following.

In a first approximation we may treat the junctions between the two mesas and the base mesa as if they were normal electrodes. In this case the only modification is the finite current through the second mesa. The charge imbalance potential generated on the first superconducting layer of the base mesa is

$$\Psi_B = \frac{F_1}{F} \psi\left(\frac{I_1}{F_1}\right) + \frac{F_2}{F} \psi\left(\frac{I_2}{F_2}\right). \quad (56)$$

The current density through the second mesa, which is kept constant, is

$$j_2 = j_0(V_2) - \sigma_0 \Psi_B, \quad (57)$$

where $j_0(V)$ is the current-voltage function (52). The voltage shift $\Delta V_2(I_1) = V_2(I_1) - \bar{V}_2$, is the difference between the voltage measured at the second mesa for fixed current I_2 , when the last barriers in the first and second mesa are in the resistive state (Fig. 7b), and the constant voltage \bar{V}_2 , when only the last barrier of the second mesa is in the resistive state (Fig. 7a). Expanding $j_0(V)$ around the voltage \bar{V}_2 as $j_0(\bar{V}_2 + \Delta V_2) = j_0(\bar{V}_2) + \Delta V_2 \sigma(\bar{V}_2)$ we obtain:

$$\Delta V_2(I_1) = \frac{\sigma_0}{\sigma(\bar{V}_2)} \frac{F_1}{F} \psi\left(\frac{I_1}{F_1}\right), \quad (58)$$

where the function $\psi(j)$ is shown in Fig. 5b.

This approximation can be improved if we take into account that the last layers in the small mesas are in the superconducting state. Then we have to include the frequency dependent density of states $u'(\epsilon)$ of these layers both in the calculation of the quasi-particle current densities and in the charge imbalance function $\alpha(\epsilon)$. Let

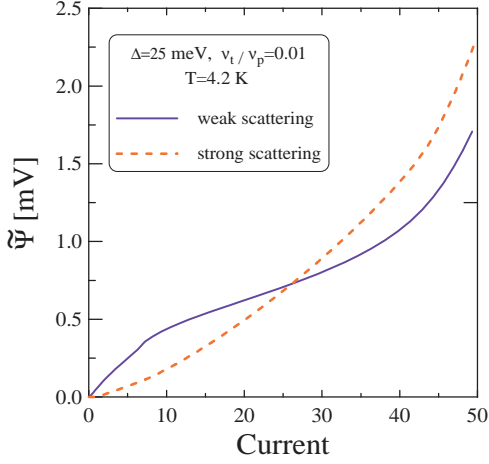


FIG. 8: Modified charge imbalance potential generated on a superconducting layer by resistive junction as function of the tunnelling current.

us denote the modified quasi-particle current by $\tilde{j}_0(V)$ and the modified charge-imbalance potential function by $\tilde{\psi}(j)$ (explicit formulas are given in the Appendix), then the voltage shift $\Delta V_2(I_1)$ measured at the second mesa is given by

$$\Delta V_2(I_1) = \frac{\sigma_0}{\tilde{\sigma}(V_2)} \frac{F_1}{F} \tilde{\psi}\left(\frac{I_1}{F_1}\right), \quad (59)$$

where $\tilde{\sigma}(V) = d\tilde{j}_0(V)/dV$. Its current dependence is proportional to the function $\tilde{\psi}(j)$, which is shown in Fig. 8. The shape of the functions $\psi(j)$, Fig. 5b and $\tilde{\psi}(j)$, Fig. 8 is similar. The curve obtained in the weak scattering limit (solid line) also has great similarity with the experimental results.⁶ The parameters used in our calculations are typical for these materials. We did not perform a fit to the experimental data, since this would require an additional parameter for the Josephson coupling. Of course all the curves end at the critical current.

D. Influence of charge-imbalance on the current-voltage curves

The same formalism can be applied to calculate the influence of charge-imbalance on the current-voltage curves in a stack of Josephson junctions. For instance, for a stack of junctions with one junction in the resistive state inside the stack (not adjoining the normal electrodes) the current is given by

$$j(V) = \tilde{j}_0(V) - 2\sigma_0\tilde{\Psi}(V), \quad (60)$$

where V is the voltage drop across this barrier. The factor 2 comes from the charge-imbalance potential created on the two superconducting layers adjoining the barrier in the resistive state.

The corresponding voltage shift for a given current j is then

$$\Delta V = 2 \frac{\sigma_0}{\tilde{\sigma}(V)} \tilde{\psi}(j), \quad (61)$$

where V is the voltage for a single junction. This shift can be measured directly as difference between the voltage of two isolated junctions in the resistive state and two neighboring resistive junctions in the stack. This generalises our results obtained with the phenomenological theory⁷ using an ohmic quasi-particle IV-curve. Note that this (very small) voltage shift in the IV-curves does not depend on the parameter a (called α in Refs.7,21 describing charge fluctuations of the superconducting condensate. These will be of importance in dynamical effects like the Josephson plasma resonance and in optical experiments.²⁸

VII. SUMMARY

In this paper we have developed a comprehensive microscopic theory for stationary nonequilibrium effects in intrinsic Josephson systems starting from a nonequilibrium Green function theory for layered d-wave superconductors. We investigated the charge-imbalance generated on a superconducting layer by current injection and derived results for the charge-imbalance distribution function and the nonequilibrium quasi-particle current between superconducting layers. The theory uses basic nonequilibrium concepts developed earlier for bulk superconductors and is applied here to layered d-wave superconductors forming a stack of Josephson junctions. Specific for layered superconductors with small tunneling rate between the layers is the confinement of charge-imbalance on single superconducting layers. Specific for d-wave superconductors with vanishing gap is the relaxation of charge-imbalance due to elastic impurity scattering, which is the dominant relaxation mechanism at low energies. In distinction to an earlier semi-phenomenological theory by the authors we considered here the energy dependence of the charge-imbalance distribution function and its relaxation, leading to non-linear current-voltage realtions.

We applied the theory to the calculation of nonequilibrium effects in current injection experiments with 4 contacts. In particular, we calculated the voltage between a normal electrode and a superconducting layer for zero current as function of the current through a second electrode. This voltage measures directly the charge imbalance potential generated on the superconducting layer. We then applied the theory to recent double-mesa experiments. Thus we were able to explain the non-linear dependence of the voltage measured at one mesa on the current through the second mesa.

The same formalism can also be applied to calculate the influence of charge imbalance on the current-voltage curves in a stack of Josephson junctions, which should

be observable as difference in the voltage between different configurations of a given number of resistive junctions in the stack. We note that the shift of the chemical potential of the condensate leads to a redistribution of the voltage between different superconducting layers, but this has no influence on the total current-voltage curves. Charge oscillations in the condensate and the resulting coupling between the layers will however be important for dynamic effects like the dispersion of the longitudinal Josephson plasma resonance and in some optical experiments. These dynamic effects will be investigated in a forthcoming publication.

Acknowledgments

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APPENDIX A: DEFINITION OF NON-EQUILIBRIUM GREEN FUNCTIONS FOR LAYERED SUPERCONDUCTORS

Following the standard definitions^{17,18} for non-equilibrium Green functions for superconductors we define larger and lesser Green functions for layers n and n' ($\vec{k} = -k$) in Nambu space by

$$\hat{G}_{nn'}^>(\vec{k}, t_1; \vec{k}', t_2) = -i \begin{pmatrix} \langle c_{n\vec{k}\uparrow}(t_1) c_{n'\vec{k}'\uparrow}^\dagger(t_2) \rangle & \langle c_{n\vec{k}\uparrow}(t_1) c_{n'\vec{k}'\downarrow}(t_2) \rangle \\ -\langle c_{n\vec{k}\downarrow}^\dagger(t_1) c_{n'\vec{k}'\uparrow}^\dagger(t_2) \rangle & -\langle c_{n\vec{k}\downarrow}^\dagger(t_1) c_{n'\vec{k}'\downarrow}(t_2) \rangle \end{pmatrix}, \quad (\text{A1})$$

$$\hat{G}_{nn'}^<(\vec{k}, t_1; \vec{k}', t_2) = +i \begin{pmatrix} \langle c_{n'\vec{k}'\uparrow}^\dagger(t_2) c_{n\vec{k}\uparrow}(t_1) \rangle & \langle c_{n'\vec{k}'\downarrow}(t_2) c_{n\vec{k}\uparrow}(t_1) \rangle \\ -\langle c_{n'\vec{k}'\uparrow}^\dagger(t_2) c_{n\vec{k}\downarrow}^\dagger(t_1) \rangle & -\langle c_{n'\vec{k}'\downarrow}(t_2) c_{n\vec{k}\downarrow}^\dagger(t_1) \rangle \end{pmatrix}, \quad (\text{A2})$$

from which we obtain the retarded advanced and Keldysh function by

$$\begin{aligned} \hat{G}^R &= \Theta(t_1 - t_2)(\hat{G}^> - \hat{G}^<), \\ \hat{G}^A &= -\Theta(t_2 - t_1)(\hat{G}^> - \hat{G}^<), \\ \hat{G}^K &= \hat{G}^> + \hat{G}^<. \end{aligned} \quad (\text{A3})$$

For the different components in Nambu space the following notation is commonly used:

$$\hat{G} = \begin{pmatrix} G & F \\ -F^\dagger & \bar{G} \end{pmatrix}. \quad (\text{A4})$$

For the average diagonal Green functions with $n' = n$ only one index and one k-vector will be used. For these

functions we introduce a Fourier transform with respect to the time-difference $\tau := t_1 - t_2$ keeping the central time $t := (t_1 + t_2)/2$ fixed:

$$\hat{G}(\vec{k}, t, \epsilon) = \int d\tau \hat{G}(\vec{k}, t + \frac{\tau}{2}, t - \frac{\tau}{2}) e^{i\epsilon\tau}. \quad (\text{A5})$$

Finally we introduce the quasi-classical approximation by integrating over the energy $\xi_k = \epsilon_k - \mu$ keeping the direction \hat{k} of the momentum fixed:

$$\hat{g}(\hat{k}, t, \epsilon) = \begin{pmatrix} g & f \\ -f^\dagger & \bar{g} \end{pmatrix} = \frac{i}{\pi} \int d\xi_k \hat{G}(\vec{k}, t, \epsilon). \quad (\text{A6})$$

APPENDIX B: DERIVATION OF KINETIC EQUATIONS

In our model the equations of motion for the Keldysh Green function on layer n in Nambu space are given by:

$$\begin{aligned} i \frac{\partial}{\partial t_1} \tau_3 \hat{G}_n^K(\vec{k}, t_1, t_2) - \hat{H}(\vec{k}, t_1) \hat{G}_n^K(\vec{k}, t_1, t_2) \\ - \{ \hat{\Sigma}^R \hat{G}^K + \Sigma^K G^A \} &= 0, \\ -i \frac{\partial}{\partial t_2} \hat{G}_n^K(\vec{k}, t_1, t_2) \tau_3 - \hat{G}_n^K(\vec{k}, t_1, t_2) \hat{H}(\vec{k}, t_2) \\ - \{ \hat{G}^R \hat{\Sigma}^K + \hat{G}^K \Sigma^A \} &= 0, \end{aligned} \quad (\text{B1})$$

where

$$\hat{H}(\vec{k}, t) = -e\phi_n(t) + \xi_k - \hat{\Delta}_k(t), \quad (\text{B2})$$

with the electrical scalar potential $\phi_n(t)$, the charge of the electron ($-e$), and the order parameter matrix

$$\hat{\Delta}_k(t) = \begin{pmatrix} 0 & \Delta_k(t) \\ -\Delta_k^*(t) & 0 \end{pmatrix}. \quad (\text{B3})$$

Here $\Delta_k(t) = \Delta_k e^{i\chi_n(t)}$ has a time-dependent phase. The constant amplitude Δ_k of the order parameter is equal on each layer and has d-wave symmetry. In order to eliminate the time-dependent phase in the order parameter we make a gauge transformation $c_{nk}(t) = \tilde{c}_{n,k}(t) \exp(i\chi_n(t))$. After this gauge transformation the new Green function \tilde{G} fulfills an equation of motion with $\hat{H}(\vec{k}, t) = -e\Phi_n(t) + \xi_k - \hat{\Delta}_k$, the gauge invariant scalar potential $\Phi_n(t) = \phi_n(t) - \hbar\dot{\chi}_n(t)/2e$, and a real time-independent order parameter.

The symbol $\{\hat{\Sigma}\hat{G}\}$ denotes a convolution in time space of the self-energy and the Green function:

$$\{AB\}(t_1, t_2) = \int dt_3 A(t_1, t_3) B(t_3, t_2). \quad (\text{B4})$$

The self-energy, which will be discussed in detail later, contains random impurity scattering within the layer and tunnelling to the neighboring layers. The latter will be treated in second order in the tunnelling matrix element.

A kinetic equation is obtained subtracting the two equations, introducing a central time $t := (t_1 + t_2)/2$ and taking the Fourier transform with respect to the time-difference $\tau := t_1 - t_2$:

$$\begin{aligned} & \left(\frac{i}{2} \frac{\partial}{\partial t} + \epsilon\right) \tau_3 \hat{G}^K + \left(\frac{i}{2} \frac{\partial}{\partial t} - \epsilon\right) \hat{G}^K \tau_3 - \{\hat{H} \hat{G}^K - \hat{G}^K \hat{H}\} \\ &= \{\hat{\Sigma}^R \hat{G}^K + \hat{\Sigma}^K \hat{G}^A - \hat{G}^R \hat{\Sigma}^K - \hat{G}^K \hat{\Sigma}^A\}. \end{aligned} \quad (B5)$$

In this equation the kinetic energy ξ_k drops out and we can perform the integration over ξ_k keeping the direction \hat{k} of the momentum fixed. We then obtain the kinetic equation for the quasi-classical Green functions

$$\begin{aligned} & \left(\frac{i}{2} \frac{\partial}{\partial t} + \epsilon\right) \tau_3 \hat{g}^K + \left(\frac{i}{2} \frac{\partial}{\partial t} - \epsilon\right) \hat{g}^K \tau_3 \\ & - \{\hat{h}(\hat{k}, t) \hat{g}^K - \hat{g}^K \hat{h}(\hat{k}, t)\} \\ &= \{\hat{\sigma}^R \hat{g}^K + \hat{\sigma}^K \hat{g}^A - \hat{g}^R \hat{\sigma}^K - \hat{g}^K \hat{\sigma}^A\} \end{aligned} \quad (B6)$$

with

$$\begin{aligned} \hat{h}(\hat{k}, t) &= -e\Phi_n(t) - \hat{\Delta}(\hat{k}), \\ \hat{\Delta}(\hat{k}) &= \begin{pmatrix} 0 & \Delta(\hat{k}) \\ -\Delta(\hat{k}) & 0 \end{pmatrix}. \end{aligned} \quad (B7)$$

The curly brackets denote a convolution in time and frequency space:

$$\{AB\}(t, \epsilon) = e^{i(\partial_t^A \partial_\epsilon^B - \partial_\epsilon^A \partial_t^B)/2} A(t, \epsilon) B(t, \epsilon). \quad (B8)$$

The equation (B6) is the starting point of our calculations.

APPENDIX C: COUPLING BETWEEN TWO SUPERCONDUCTING LAYERS

a) Tunnelling between two superconducting layers with the barrier in the superconducting state

Here we have $\Omega_{n,n'} = 0$ but a finite constant phase difference $\varphi := \gamma_{n,n'}^0$, and we obtain for the tunnelling self-energy

$$\begin{aligned} \hat{\sigma}_t^R(\hat{k}, \epsilon)_{n,n'} &= -i\langle \nu_t g^R(\epsilon) \rangle' \tau_3 + \sin \varphi \langle \nu_t f^R(\epsilon) \rangle' \tau_1 \\ &+ \cos \varphi \langle \nu_t f^R(\epsilon) \rangle' \tau_2, \end{aligned} \quad (C1)$$

$$\begin{aligned} \hat{\sigma}_t^K(\hat{k}, \epsilon) &= -2i\langle \nu_t u'(\epsilon) \beta'(\epsilon) \rangle' \tau_3 - 2i\langle \nu_t u'(\epsilon) \alpha'(\epsilon) \rangle' \tau_0 \\ &+ 2\sin \varphi \langle \nu_t v'(\epsilon) \beta'(\epsilon) \rangle' \tau_1 \\ &+ 2\cos \varphi \langle \nu_t v'(\epsilon) \beta'(\epsilon) \rangle' \tau_2 \\ &+ 2\cos \varphi \langle \nu_t w'(\epsilon) \alpha'(\epsilon) \rangle' \tau_1 \\ &- 2\sin \varphi \langle \nu_t w'(\epsilon) \alpha'(\epsilon) \rangle' \tau_2. \end{aligned} \quad (C2)$$

For the contribution from tunnelling to the scattering term in the kinetic equation we then find:

$$\begin{aligned} I_{11}^{ts} - I_{22}^{ts} &= -8i\langle \nu_t u'(\epsilon) u(\epsilon) \alpha(\epsilon) \rangle' + 4i\langle \nu_t u'(\epsilon) \alpha'(\epsilon) u(\epsilon) \rangle', \\ J_{12}^{ts} - J_{21}^{ts} &= 8i\langle \nu_t u'(\epsilon) \alpha'(\epsilon) v(\epsilon) \rangle'. \end{aligned} \quad (C3)$$

Note that all the contributions depending on the phase difference φ drop out. The remaining terms vanish if we neglect terms of order $\nu_t \alpha$ which are of higher order in the tunnelling probability.

b) Tunnelling between two superconducting layers with the barrier in the resistive state.

In this case also the anomalous Green functions contribute to the dc current. As this contribution involves the coherent part of the tunnelling matrix element and will be small in the limit of large McCumber parameters, we neglect it in the following. However, we have to take into account the frequency dependent density of states $u(\epsilon), u'(\epsilon)$ on both superconducting layers. We want to apply the theory to calculate the charge-imbalance α on a superconducting layer n which is coupled on one side to a superconducting layer n' in a stack with barriers in the resistive state and on the other side to a superconducting layer with the barrier in the superconducting state. As the layer n' is between two barriers in the resistive state with equal quasi-particle current we can neglect the charge-imbalance α' on this layer. Then we obtain the contribution to the scattering term from tunnelling between n and n' :

$$\begin{aligned} I_{11}^{tn} - I_{22}^{tn} &= -4i\langle \nu_t (u'(\epsilon - v) - u'(\epsilon + v)) u(\epsilon) \beta(\epsilon) \rangle' \\ &+ 4i\langle \nu_t (u'(\epsilon - v) \beta'(\epsilon - v) - u'(\epsilon + v) \beta'(\epsilon + v)) u(\epsilon) \rangle' \\ &- 4i\langle \nu_t (u'(\epsilon - v) + u'(\epsilon + v)) u(\epsilon) \alpha(\epsilon) \rangle', \\ J_{12}^{tn} - J_{21}^{tn} &= 4i\langle \nu_t v(\epsilon) \rangle (u'(\epsilon - v) \beta'(\epsilon - v) \\ &- u'(\epsilon + v) \beta'(\epsilon + v)) \rangle'. \end{aligned} \quad (C4)$$

For the charge-imbalance function $\alpha(\epsilon)$ on the superconducting layer n we then find:

$$\begin{aligned} & 2\nu_p [u(\epsilon) \langle u(\epsilon) \alpha(\epsilon) \rangle - \langle u(\epsilon) \rangle u(\epsilon) \alpha(\epsilon)] \\ & - 2\nu_p \frac{\Delta v(\epsilon)}{\bar{\epsilon}} \langle u(\epsilon) \alpha(\epsilon) \rangle \\ &= -\left(u(\epsilon) - \frac{\Delta}{\bar{\epsilon}} v(\epsilon)\right) \langle \nu_t(\hat{k}, \hat{k}') [u'(\epsilon - v)(\beta'(\epsilon - v) - \beta(\epsilon)) \\ &+ u'(\epsilon + v)(\beta(\epsilon) - \beta'(\epsilon + v))] \rangle'. \end{aligned} \quad (C5)$$

The formulas for the current densities obtained from Eq.(51) now read in (symmetrised form)

$$j^\beta(V) = \frac{e}{2\hbar} N(0) \nu_t \int_{-\infty}^{+\infty} d\epsilon \langle u(\epsilon) \rangle \langle u'(\epsilon + eV) \rangle \quad (C6)$$

$$\begin{aligned} & + u'(\epsilon - eV) (\beta_0(\epsilon + eV) - \beta_0(\epsilon - eV)) =: \tilde{j}_0(V), \\ j^\alpha(V) &= -\frac{e}{\hbar} N(0) \nu_t \int_{-\infty}^{+\infty} d\epsilon \tilde{\alpha}(\epsilon) \langle u'(\epsilon + eV) \rangle \\ & + u'(\epsilon - eV) =: -\sigma_0 \tilde{\Psi}(V), \end{aligned} \quad (C7)$$

with the charge imbalance function $\tilde{\alpha}(\epsilon) = \langle u(\epsilon) \alpha(\epsilon) \rangle$ given by

$$\begin{aligned} \tilde{\alpha}(\epsilon) &= \frac{\nu_t \langle R(\epsilon) \rangle}{2\nu(\epsilon)} \langle u'(\epsilon + eV) + u'(\epsilon - eV) \rangle \\ & \quad \langle \beta_0(\epsilon + eV) - \beta_0(\epsilon - eV) \rangle. \end{aligned} \quad (C8)$$

The current equation for $j^\alpha(V)$ defines a modified charge-imbalance potential $\tilde{\Psi}(V)$, from which we obtain the function $\tilde{\psi}(j) = \tilde{\Psi}(V)$ using the current-voltage relation $j = \tilde{j}_0(V)$. This function is shown in Fig. 8.

APPENDIX D: CHARGE IMBALANCE OF QUASI-PARTICLES

In order to make contact with the traditional theory of charge imbalance let us summarise the basic definitions and concepts of charge-imbalance as introduced by Tinkham and Clarke^{10,11,31,32}.

In the BCS theory the total charge (in units of the electron charge, factor 2 from spin) is

$$Q = 2 \sum_k v_k^2 + (u_k^2 - v_k^2) f_k =: Q^c + Q^*, \quad (D1)$$

where $u_k^2 = \frac{1}{2}(1 + \xi_k/E_k)$, $v_k^2 = (1 - \xi_k/E_k)/2$ are the usual coherence factors, $\xi_k = \epsilon_k - \mu$, $E_k = \sqrt{\xi_k^2 + \Delta_k^2}$ is the quasi-particle excitation energy. The first term is the condensate charge Q^c , the second term is the quasi-particle charge Q^* , f_k is the quasi-particle distribution function. In equilibrium $f_k = 1/(\exp(E_k/T) + 1)$ and hence Q^* vanishes (for particle-hole symmetry).

A nonequilibrium state can be described by a shift $\delta\mu$ of the chemical potential and a change of the distribution function f_k . A shift of the chemical potential leads to a shift of the excitation energy in k-space, $\xi_k \rightarrow \epsilon_k - \mu + \delta\mu$ and hence to a change of the condensate charge by:

$$\delta Q^c = 2N(0)\delta\mu, \quad (D2)$$

where $N(0)$ is the density of states for one spin direction at the Fermi energy.

The quasi-particle charge can also be written as

$$Q^* = 2 \sum_k q_k f_k = \sum_k q_k (f_{k>} - f_{k<}), \quad (D3)$$

where $q_k = \xi_k/E_k$ can be considered as charge of a quasi-particle with momentum \vec{k} . For each quasi-particle state with momentum $\vec{k} = \vec{k}_{>}$ and $\xi_{k_{>}} > 0$ (electron-like quasi-particle) exists a quasi-particle state with momentum $\vec{k} = \vec{k}_{<}$ and $\xi_{k_{<}} = -\xi_{k_{>}} < 0$ (hole-like quasi-particle) with the same excitation energy $E_k > 0$ and direction \hat{k} . Thus Q^* depends on the difference in the number of electron- and hole-like quasi-particles. While $f_{k_{>}} - f_{k_{<}}$ describes charge-imbalance, the combination $f_{k_{>}} + f_{k_{<}}$, for instance, enters the self-consistency equation for the gap Δ_k .

These distribution functions are introduced here for well-defined quasi-particles with infinite life-time. In a microscopic theory based on nonequilibrium Green functions these are replaced here by the frequency-dependent distribution functions $\alpha(\epsilon)$ and $\beta(\epsilon)$. In our case we find the following correspondence for $\epsilon > 0$:

$$\beta(\epsilon = E_k) = 1 - f_{k_{>}} - f_{k_{<}}, \quad (D4)$$

$$\alpha(\epsilon = E_k) = -q_k(f_{k_{>}} - f_{k_{<}}). \quad (D5)$$

Extended to the whole frequency range $\beta(\epsilon)$ becomes an odd function, $\alpha(\epsilon)$ is an even function.

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